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ALKALI-RARE GAS AND METAL HALIDE MOLECULES AS POTENTIAL VISIBLE LASERS

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FINAL REPORT FOR AFOSR CONTRACT ▓▓▓▓▓

Period: October 1, 1979 through September 30, 1980

"Alkali-Rare Gas and Metal Halide
Molecules as Potential Visible Lasers"

Prepared for

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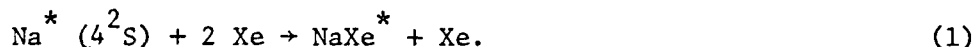
SPONSORSHIP (CONTRACT #79-0138).

1. Both NaI and KI have been photodissociated using an ArF excimer laser - the blue $NaXe^*$ excimer band has been observed in NaI-Xe mixtures.

2. The $NaXe^*$ threebody formation rate has been determined to be $k_f \sim 10^{-32}$ $cm^6 - sec^{-1}$.

3. The laser driven photochemical reaction:

$$NaI + O_2 + 2h\nu \rightarrow NaO^* + products \rightarrow NaO_2 + I_2^-$$

has been observed.

4. The blue-green emission band of IF^* (iodine-monofluoride) has been identified for the first time and the IF^* formation kinetics (under e-beam excitation) have been determined.

5. Lasing has been obtained on five different lines of the IF^* blue-green band with discharge excitation.

6. The gain and transient absorption spectra of IF and ICℓ have been measured.

7. The radiative lifetimes and quenching rate constants (for various gases) have been measured for both IF^* and $ICℓ^*$.

8. $PbCℓ_2$ and CdI_2 have been photodissociated (again using an ArF laser) and the visible emission spectra of $PbCℓ^*$ and CdI^* were recorded.

9. Preliminary gain measurements have been conducted for CdI^* using a tunable dye laser. No gain was observed between 625.0 and 632.0 nm for CdI_2 pressures ranging from 1 to 50 Torr.

10. Six papers have resulted from this research effort and have either been published or have been submitted for publication.

I. INTRODUCTION

The major goal of this research effort is to identify those members of the alkali-rare gas and metal-halide molecular families which show promise as efficient, powerful and practical visible lasers. Laser photodissociation of polyatomic molecules and self-sustained discharge pumping are the primary tools being used to demonstrate lasing from these molecules.

II. RESULTS FOR THE ALKALI-RARE GAS MOLECULES

Our initial experiments have focussed on sodium and Fig. 1 is a partial energy level diagram for the Na atom. Our goal was to produce the NaXe* molecule and investigate its potential as a tunable blue laser. This has been accomplished by photodissociation of the NaI molecule (using an ArF excimer laser) in NaI-Xe mixtures. The energy of an ArF laser photon(193 nm) is $\sim$ 6.43 eV which is sufficient to break the NaI bond and leave the Na atom in the 4^2S state. Now, if the Xe pressure is sufficiently high, then the Na* (4^2S) atom, rather than radiatively decaying to the 3^2P levels, will form a NaXe* molecule in the 3-body collision:

$$Na^* (4^2S) + 2\ Xe \rightarrow NaXe^* + Xe. \tag{1}$$

Subsequently, the NaXe* molecule will radiate to its dissociative ground state, giving rise to an emission band in the blue at 440 nm. We have observed this band using the experimental apparatus diagrammed in Fig. 2. Also, since the NaXe ground state is dissociative (repulsive) as shown in Fig. 1, the NaXe molecule breaks up after radiating, NaI is then reformed and the whole process is repeated. In other words, the photodissociative process is cyclical. This has also been observed in that no decrease in the NaXe blue fluorescence (from shot to shot) has been observed to date.

For the experiments performed thus far, [Na] $\sim 10^{16}$ cm^{-3} and [Xe] $\sim 1.6 \cdot 10^{19}$ cm^{-3}. For these densities, experiments have shown that most of the absorbed laser energy appears in the Na resonance lines.
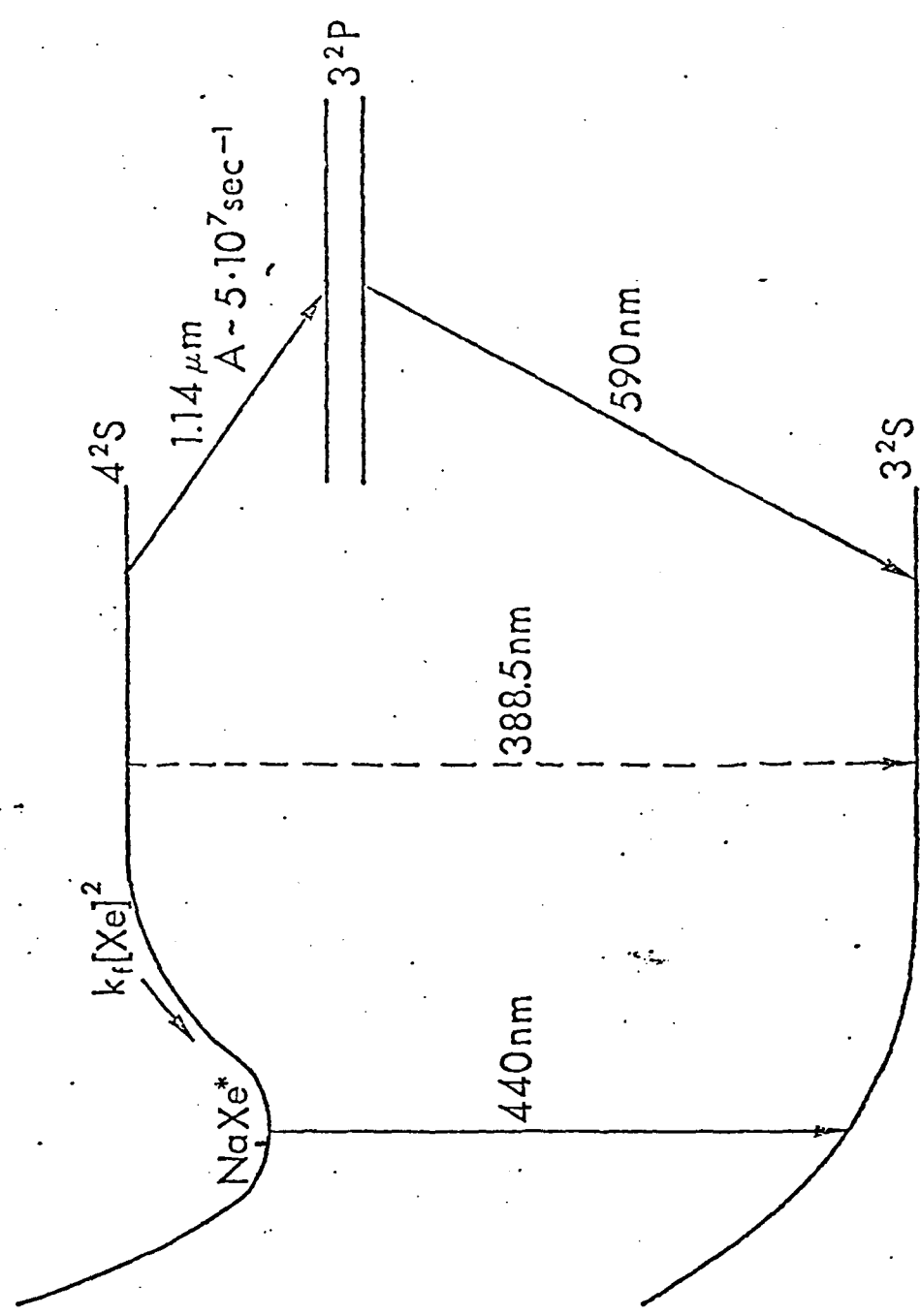
Fig. 1. Partial energy level diagrams for the Na atom and the NaXe* excimer. The Na(4^2S $\to$ 3^2S) transition is forbidden and is denoted by the dashed line. Also note that formation of the excimer by three-body collisions competes with ...
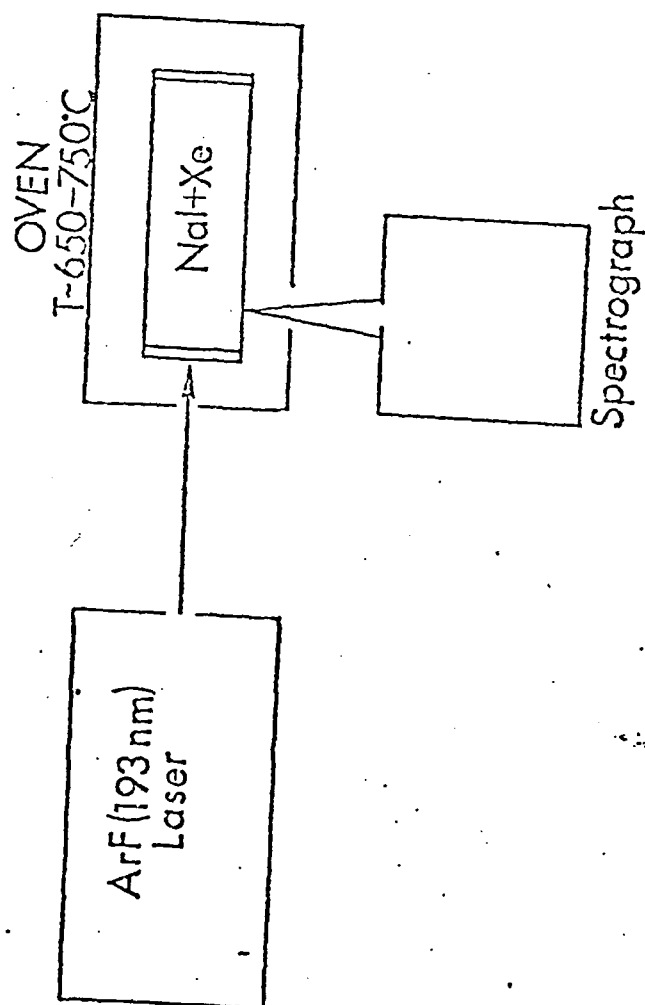
Fig. 2. Experimental apparatus used in the photodissociation of NaI to form the NaXe* excimer.

Therefore, simple rate equation calculations reveal that the NaXe excimer formation rate constant, k_f, is $\sim 10^{-32}$ cm^6-sec^{-1} or smaller than expected from the literature.

The low measured value of k_f by itself is not sufficient to eliminate NaXe* from consideration as a viable laser candidate. However, as shown in Fig. 1, the Na(4^2S) state is optically connected to the 3^2P level via a fast infra-red transition at 1.14 μm. Unfortunately, the large transition probability of this line makes it extremely difficult to build up a large NaXe* population unless the Xe pressure is made unreasonably large. Stated another way, the branching ratio for NaXe* production:

$$\frac{k_f [Xe]^2}{\tau_{IR}^{-1} + k_f [Xe]^2} = 50\%$$

(where $k_f \sim 10^{-32}$ cm^6 - sec^{-1} and τ_{IR} is the 20 ns radiative lifetime of the 4^2S $\rightarrow 3^2$P transition) when the Xe density, [Xe], is $7 \cdot 10^{19}$ cm^3. This extremely large Xe pressure (2200 Torr at room temperature) would place severe constraints on any practical system and therefore our research efforts have shifted to the metal halide molecules.

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III. GROUP IIB METAL-HALIDE EXPERIMENTS

Recently, then, we initiated a series of experiments designed to determine the feasibility of obtaining laser emission from CdI, PbCℓ and ZnI. These molecules were chosen because: 1) temperatures $\stackrel{<}{\sim}$ 400°C are required to obtain one Torr of the associated triatomic (CdI_2, $PbCℓ_2$ or ZnI_2); 2) the lowest electronic excited states of these molecules are strongly optically connected to ground, giving rise to visible emission bands, and 3) the lowest excited state and ground are Franck-Condon shifted which considerably reduces the threshold population inversion.

An experimental arrangement similar to that shown in Fig. 1 was used. A Suprasil quartz tube, which contained a small amount of the desired salt and 50 Torr of Ne, was placed in an oven and heated to the temperature required to obtain $\sim$ 0.1 - 50 Torr of the triatomic salt. Photodissociation was produced by focussing the output of the ArF laser and the resulting fluorescence was recorded by a spectrograph and Polaroid film. Strong visible emission has been observed from all of these molecules.

Initial experiments have been made to probe CdI^* for gain in the red where the peak emission occurs. The rectangular output beam of the ArF laser was focussed to a line inside the CdI_2 tube using a cylindrical lens. The probe laser beam, which was provided by a Chromatix pulsed tunable dye laser, was directed along the axis of the tube and coincident with the pump pulse. To insure that the probe beam and the pump focus were indeed coincident, the dye laser was first tuned to the $5d^1D_2 \rightarrow 5p^1P_1$ transition of atomic Cd at 644.0 nm. (This line appears strongly in the emission of ArF pumped CdI_2 suggesting a multi-photon process). The physical position of both beams were adjusted until absorption on this transition was observed (see Fig. 3).

Unfortunately, no gain was observed in the 625.0 to 632.0 nm spectral region for CdI_2 pressures ranging from 1 to 50 Torr. Presently, these gain measurements are being extended to CdI_2 pressures of a fraction of a Torr. In the near future, similar experiments involving ZnI_2 and $SnCl_2$ will be conducted. For all of these experiments, the photodissociation of $HgBr_2$ to produce $HgBr^*$ molecules is being used as a "reference." The reason for this is simply that the stimulated emission cross-section for the HgBr laser is well-known and is a useful yardstick in assessing the laser potential of one of the molecules mentioned earlier.
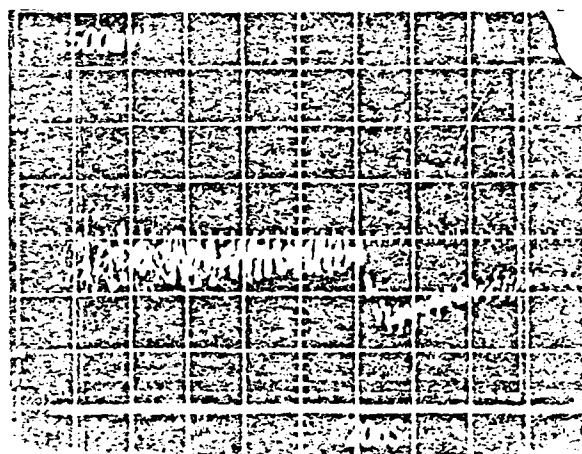
Fig. 3. Absorption observed on the $Cd(5d^1D_2 \rightarrow 5p^1P_1)$ line at 644 nm following photodissociation of CdI_2. This absorption was ·used to align the pump focus and probe laser beam.

IV. IODINE-MONOFLUORIDE

Early in the contract year, while waiting for our excimer laser to arrive, several experiments were conducted to investigate the IF (iodine-monofluoride) molecule as a potential visible laser. A small electron beam generator in our laboratory made convenient the study of the emission spectrum and formation kinetics of this molecule. From these studies:

1. the IF blue-green emission spectrum was observed and tentatively assigned to the $E \to A^3\Pi$ transition,

2. the IF(E) state radiative lifetime was estimated to be 15 ns,

3. a scheme for the formation kinetics of IF(E) under e-beam pumping conditions was developed (see the enclosed reprint).

Subsequently, lasing has been observed on IF in discharge pumped $He/Ar/NF_3/CF_3I$ gas mixes using the Lumonics TE 262-2 excimer laser as the discharge chamber. In all, lasing has been observed on five different lines ranging from 472 to 497 nm (see Fig. 4). With optimum ($\sim 35\%$) output coupling, energies on the order of 4 mJ in a ~ 30 ns FWHM pulse (~ 140 kW peak power) has been obtained.

Next, the gain and transient absorption profiles of IF were measured in discharge plasmas to determine the potential tunability of the laser. The curves, shown in Fig. 4, exhibit several interesting features. First, as expected, lasing occurs at the peaks in the gain curve. Secondly, the gain is $\gtrsim 1\% - cm^{-1}$ from 478 to 497 nm, a range of almost 200 Å. Finally, the strong absorption at 492.1 nm can be attributed to the He $(2p^1P_1)$ excited species.

Attempts to remove the absorbing species have met with some success. By using Penning ionization of Ar or excitation transfer to Ne, the small signal gain-to-absorption ratio, as well as the temporal width of the gain pulse was improved considerably.
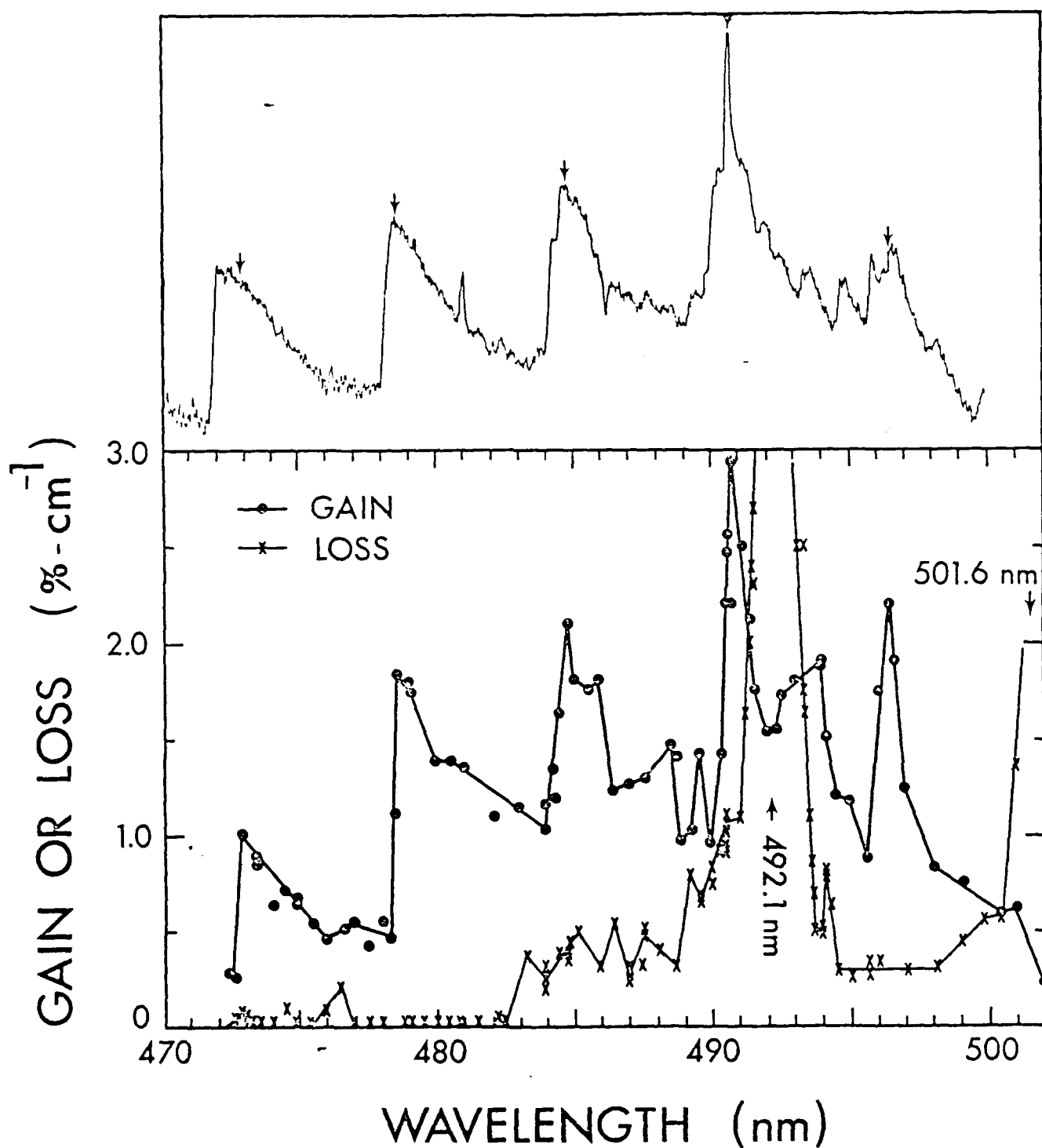
Fig. 4. (Top) Spontaneous emission spectrum of IF in discharge excited He/CF₃I/NF₃ gas mixtures. The arrows indicate those wavelengths at which lasing has been observed.

(Bottom) The gain and transient absorption spectra for IF showing strong absorption by the He background gas in the vicinity of 492 nm.

Finally, the IF(E) lifetime and quenching rate constants were measured by photodissociating IF_5 in the presence of the quenching molecule with an ArF laser. The measured lifetime of 17 ns agrees well with the estimate obtained from the e-beam data. Table I lists the measured quenching rate constants.

Similar gain and quenching measurements have been conducted for ICl and the results show a peak gain coefficient of $1.3\% - cm^{-1}$ at 431.3 nm. These results are currently being prepared for publication.

IF* QUENCHING RATE CONSTANTS

QUENCHER	$k_Q \ (cm^3-sec^{-1})$
He	$(2.2 \pm 1.0) \cdot 10^{-12}$
Ne	$(9.2 \pm 2.5) \cdot 10^{-13}$
Ar	$(1.5 \pm 0.7) \cdot 10^{-12}$
NF_3	$(6.8 \pm 1.6) \cdot 10^{-11}$
CF_3I	$(1.1 \pm 0.3) \cdot 10^{-10}$
IF_5	$(2.2 \pm 0.3) \cdot 10^{-10}$

TABLE I. The rate constants for two body quenching of IF* by various atoms and molecules.

PUBLICATIONS UNDER AFOSR SPONSORSHIP

1) Iodine-monofluoride emission spectrum and formation kinetics in electron-beam-produced plasmas.

S.B. Hutchison, J.G. Eden, and J.T. Verdeyen, Appl. Phys. Lett. $\underline{37}$, 374 (1980).

2) Multi-line (480-496 nm) discharge pumped iodine-monofluoride laser.

M.L. Dlabal, S.B. Hutchison, J.G. Eden, and J.T. Verdeyen. (To be published in Appl. Phys. Lett. (Nov. 15, 1980)).

3) 140 kW IF laser: Small signal gain and operating parameters.

M.L. Dlabal, S.B. Hutchison, J.G. Eden, and J.T. Verdeyen. (To be published in Optics Letters).

4) Gain and transient absorption profiles for the iodine-monofluoride 430 nm bands under discharge excitation.

M.L. Dlabal and J.G. Eden. (To be published in Applied Physics Letters).

5) IF* radiative lifetime and quenching rate constants by ArF photo-dissociation of IF_5.

M.L. Dlabal and J.G. Eden. (To be published).

6) Radiative lifetime and heavy particle quenching of ICl^*.

M.L. Dlabal, S.B. Hutchison and J.G. Eden. (To be published).

140 kW IF LASER: SMALL SIGNAL GAIN AND

OPERATING PARAMETERS[*]

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Abstract

Output energies in excess of 4 mJ in a $\sim$ 30 ns FWHM pulse ($\sim$ 140 kW peak power) have been obtained from a discharge pumped IF laser for a cavity output coupling of 35%. In addition, oscillation on a new transition of the IF (E $\rightarrow$ A) band at 472.7 nm has been observed. By measuring the output power of the laser for various values of output mirror transmission, the small signal gain and loss coefficients were found to be $(3.1 \pm 0.7)\%$ – cm^{-1} and $\sim 0.3\%$ – cm^{-1}, respectively.

In the past year, fluorescence[1] and lasing[2-5] on the E → A $^3\Pi$ blue-green band[6-3] of the IF molecule have been reported. Both electron beam and discharge excitation of rare gas (usually Ar or He), CF_3I and NF_3 gas mixtures have proven successful in producing stimulated emission in the vicinity of 491 nm. The room temperature operation of this interhalogen laser and its reliance on technology previously developed for the rare gas-halide laser family make it potentially attractive for several applications requiring coherent radiation in the 450-500 nm spectral region.

The observation of IF laser output energies in excess of 4 mJ in a ∿ 30 ns FWHM pulse (∿ 140 kW peak power) is described here. This represents an order of magnitude improvement in IF pulse energy over previously published results.[2,4] Also, in addition to the previously observed[3,4] lines at 478.7, 484.7, 490.7 and 496.5 nm, lasing has been obtained on a new transition of the IF (E → A) band at 472.7 nm. Finally, measurements of the laser output power versus the transmission of the cavity output coupling mirror have been made. The results of these measurements were examined theoretically by numerically solving the differential equations that describe the temporal evolution of the intracavity photon flux and the IF population inversion. From this analysis, it is concluded that the small signal gain and loss coefficients at 491 nm are $(3.1 \pm 0.7)\%$ - cm^{-1} and ∿ 0.3% - cm^{-1}, respectively.

The experimental apparatus has been described previously[4] and will only be briefly reviewed here. Pumping of the He, CF_3I and NF_3 gas mixtures used in these experiments was provided by a commercially available (Lumonics) UV - preionized discharge system. This device has a gain length of 100 cm and produces an excited volume of ∿ 0.4ℓ. The stable optical cavity was composed of two dielectric - coated mirrors, separated by 150 cm and mounted internally to minimize intracavity losses. One

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mirror was highly reflecting (R > 97%) throughout the visible while the output coupler was chosen from one of several mirrors with transmissions at 491 nm ranging from $\sim$ 0.1 to 92%. The transmission profile of each mirror over the wavelength region of interest was measured by a Cary 14 spectrophotometer. All of the gases for these experiments (research grade rare gases and technical grade CF_3I and NF_3) were used as supplied by the manufacturer.

The spontaneous emission and laser spectra of the discharge produced plasmas were recorded either by a 0.5 m Hilger-Engis spectrograph (in first or second order) and Polaroid film or by an optical multichannel analyzer. Spectral traces were made from photographic negatives using a Joyce-Loebl microdensitometer. Finally, measurements of the laser pulse power were conducted with a calibrated S-20 surface photodiode and a Tektronix 7104 oscilloscope.

Figure 1 illustrates the emission spectra of the IF laser near (bottom of Fig. 1) and well above (top) threshold. For both spectra, the gas mixture composition was $\sim$ 1540 Torr He, 3 Torr NF_3, and 1 Torr CF_3I. The bottom trace was obtained with an optical cavity composed of the high reflector and the quartz flat, and was recorded after the discharge had been fired roughly 50 times so that the IF laser output had fallen to a point near threshold. Amplified spontaneous emission (ASE) is clearly present at $\lambda \sim$ 491 nm. Also, distortion of the spontaneous emission spectrum between 485 and 498 nm indicates the existence of significant background absorption in this spectral region. Since this has not been observed in the IF spectra of electron beam pumped NF_3 and CF_3I mixtures with an _argon_ buffer[1], it is likely that this absorption is

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due to He excited states. In contrast, the vibrational bands peaking at
$\sim$ 472 and 479 nm appear to be relatively free of background absorption
and emission.

Although the IF molecule was observed to lase at five wave-
lengths (472.7, 478.7, 484.7, 490.7 and 496.5 nm), only the 485 and
491 nm lines are shown in the higher resolution trace at the top of
Fig. 1. (Stimulated emission on the 472.7 nm transition has not
previously been reported). This laser spectrum was obtained with a
high-Q optical cavity (two R > 97% mirrors) and in order to resolve
several of the vibrational-rotational lines, the emission profile was
recorded in second order. Also, the resolution of the detection
system was $\sim$ 0.1 and 0.32 nm for the upper and lower emission spectra,
respectively.

Lasing was observed only when NF_3 was the fluorine atom donor
although several other molecules, such as F_2 and COF_2, were also investi-
gated. Diluents other than He (Ne and Ne/N_2 mixtures, in particular) were
also tried without success. Maximum laser output energy was obtained for
the 99.7% He, 0.2% NF_3 and $\sim$ 0.1% CF_3I ($p_{TOTAL} \sim$ 1545 Torr) gas mixture
mentioned earlier. To date, single pulse energies in excess of 4 mJ have
been extracted from the laser in pulses of typically 25-30 ns FWHM, for a
peak laser power of $\sim$ 140 kW. Optimum cavity output coupling was found
to be $\sim$ 35% and the output beam was observed to rectangular in cross-
section with an area of $\sim$ 2 cm^2. Although the present efficiency of the
IF laser is low ($\sim 10^{-2}$%), a more complete understanding of the kinetics
of the discharge pumped laser should lead to much better performance.

The rapid deterioration of the IF laser output energy for a
static gas fill is shown in Fig. 2. The lifetime of any particular gas
mixture was found to be sensitive to the care given to passivation of

the discharge chamber. In fact, both the gas fill lifetime and the peak

pulse energy of the laser slowly improve with time. Consequently, since

the data shown in the Figure was taken, the half-power lifetime of the

laser has been extended to 50-75 shots. As pointed out in ref. 9 for

another NF_3 containing gas laser (KrF), the decay of the IF output energy

is believed to be largely due to a rapid buildup of N_2F_2 in the cavity.

Also, the gradual depletion of CF_3I through the generation of IF_5 or IF_7

limits the useful lifetime of a gas mix. Fortunately, circulation of the

gas mixture through a simple cold trap should be sufficient to eliminate

these contaminants.

Figure 3 depicts the variation of the IF laser output power

with the partial pressure of NF_3. The concentration of CF_3I in the gas

mixture was found to be even more critical for proper operation of the

laser. Maximum output was obtained for a CF_3I partial pressure of 1.0 -

1.25 Torr but fell to 50% for p_{CF_3I} = 0.8 and 1.4 Torr.

The peak output power of the IF laser was measured for output

mirror transmissions of $\sim$ 0.1, 35, 65, 72 and 92%. As shown in Fig. 4,

the laser power is relatively insensitive to the cavity output coupling

over the interval 35-65%. To estimate the small signal gain and loss

coefficients for this laser, two separate approaches were used. The

analysis developed by Rigrod[10] for high gain lasers and subsequently

modified by Champagne[11] was invoked initially. The solid line in Fig. 4,

which represents the best fit of the equation in ref. 11 to the data,

yields a small signal gain coefficient of g_o = (1.8 ± 0.3)% - cm^{-1}.

However, Rigrod's analysis was developed for quasi-CW lasers where the

laser pulse length is long compared to the upper state radiative lifetime.

This condition is not met in these experiments (τ_{IF}* = 17 ns)[12] and so

it was necessary to solve numerically (for various values of g_o) the

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·the coupled differential equations which describe the time evolution of the intracavity photon flux and the IF population inversion. The dotted curve of Fig. 4 illustrates the solution that most closely matches the data. Accounting for the uncertainty in the laser power measurements (± 10%), the small signal gain and loss coefficients were found to be $g_o = (3.1 \pm 0.7)\% - cm^{-1}$ and $\alpha \sim 0.3\% - cm^{-1}$, respectively. This value for g_o is in agreement with that reported by DeYoung.[15]

Also, from the analysis given above, the laser saturation intensity was estimated to be $I_{sat} \sim 200$ kW $- cm^{-2}$. Now, I_{sat} can be expressed as:

$$I_{sat} = \frac{h\nu}{\sigma_{se} \, \tau_{eff}} \tag{1}$$

where σ_{se} is the stimulated emission cross-section for the IF (E → A) band and τ_{eff} is the effective lifetime (i.e., includes quenching collisions) of the E state for the optimum gas mixture used in these experiments. Taking τ_{eff} to be ~ 10 ns,[12] then $\sigma_{se} \sim 2 \cdot 10^{-16}$ cm^2 or 1.4Å^2.

In summary, 140 kW laser pulses have been obtained on the blue-green E → A band of the IF molecule. Moreover, a new transition at 472.7 nm has been observed to lase. The present, low efficiency of this laser is partially due to the fact that the time available for buildup of the optical pulse is severely limited. It is likely, therefore, that oscillator-amplifier experiments or extension of the discharge current pulse width will result in a significant increase in output energy from the IF laser, perhaps to the tens of millijoules level.

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[4] M.L. Dlabal, S.B. Hutchison, J.G. Eden and J.T. Verdeyen, "Multi-Line (480-496 nm) Discharge Pumped IF* Laser," Appl. Phys. Lett. (to be published).

[5] R.J. DeYoung, "Lasing Characteristics of Iodine-monofluoride," Appl. Phys. Lett. (to be published).

[6] The identity of the states responsible for the IF-blue-green band is presently unresolved. There is some spectroscopic evidence (see ref. 7) for an $A'(^3\Pi_{2u})$ level which, lying in proximity to the $A^3\Pi_1$ state reported in ref. 8, may be the lower energy level for this transition. Consequently, Kompa and co-workers (ref. 2) attribute this emission to the $D' \rightarrow A'$ band of the molecule.

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      Excimer Lasers Using NF_3," Appl. Phys. 13, 1 (1977).

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      Phys. 36, 2487 (1965).

[11] L.F. Champagne, "Efficient Operation of the Electron Beam Pumped
      XeCl," Appl. Phys. Lett. 33, 523 (1978).

[12] Measurements of the IF^* radiative lifetime and collisional
      quenching rate constants are in progress and the results will
      be reported elsewhere.

Fig. 1. Densitometer tracings of the spontaneous emission (bottom)
and laser (top) spectra for the IF molecule in discharge excited
99.7% He, 0.2% NF_3 and 0.1% CF_3I (p_{TOTAL} $\sim$ 1545 Torr) gas mixtures.
Although five laser lines have been observed, only the 491 and 485 nm
transitions (recorded in second order) are shown in order to illustrate
the richness of the spectra. The spectrograph resolution was $\sim$ 0.1
and 0.32 nm for the top and bottom traces, respectively. For both
spectra, the vertical scale is linear in optical density.

Fig. 2. Decay of the IF laser peak output power from shot-to-shot for
a static gas fill. The gas mixture used for these experiments is again
$\sim$ 1540 Torr He, 3 Torr NF_3 and 1 Torr CF_3I. The decline in power is
probably due to buildup of N_2F_2 in the discharge chamber and conversion
of CF_3I into IF_5.

Fig. 3. Variation of the IF laser power output with the partial pressure
of NF_3 in 1540 Torr He, p_{NF_3}, and 1 Torr CF_3I gas mixtures.

Fig. 4. Dependence of the IF laser peak power on the transmission of
the output mirror at λ = 491 nm. The solid line represents the best
fitting of Rigrod's analysis (cf. refs. 10 and 11) to the data. The
numerical solution for the coupled differential equations described in
the text (where g_o = 3.1% $-$ cm^{-1} and α $\sim$ 0.3% $-$ cm^{-1}) is indicated by the
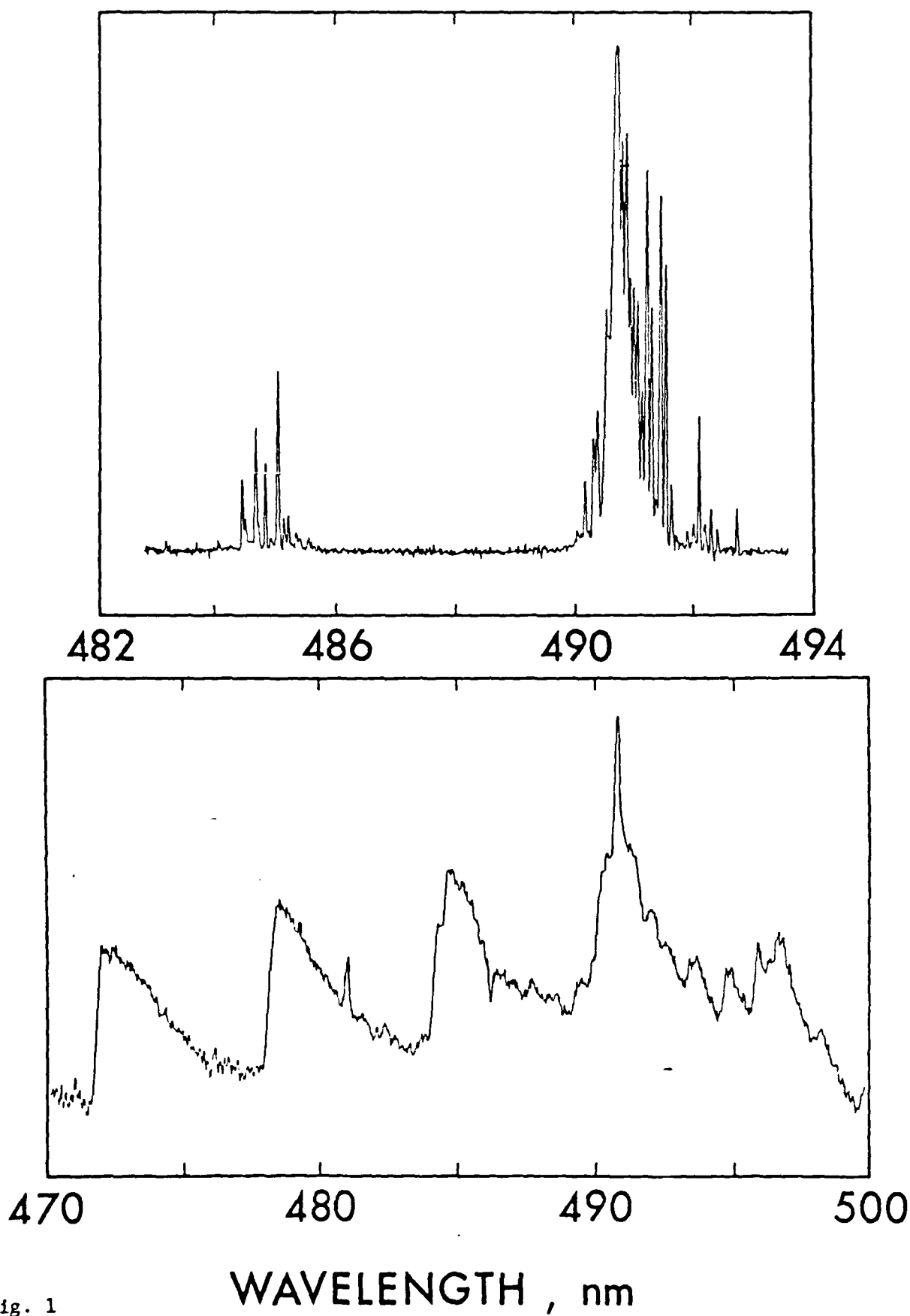dotted curve.
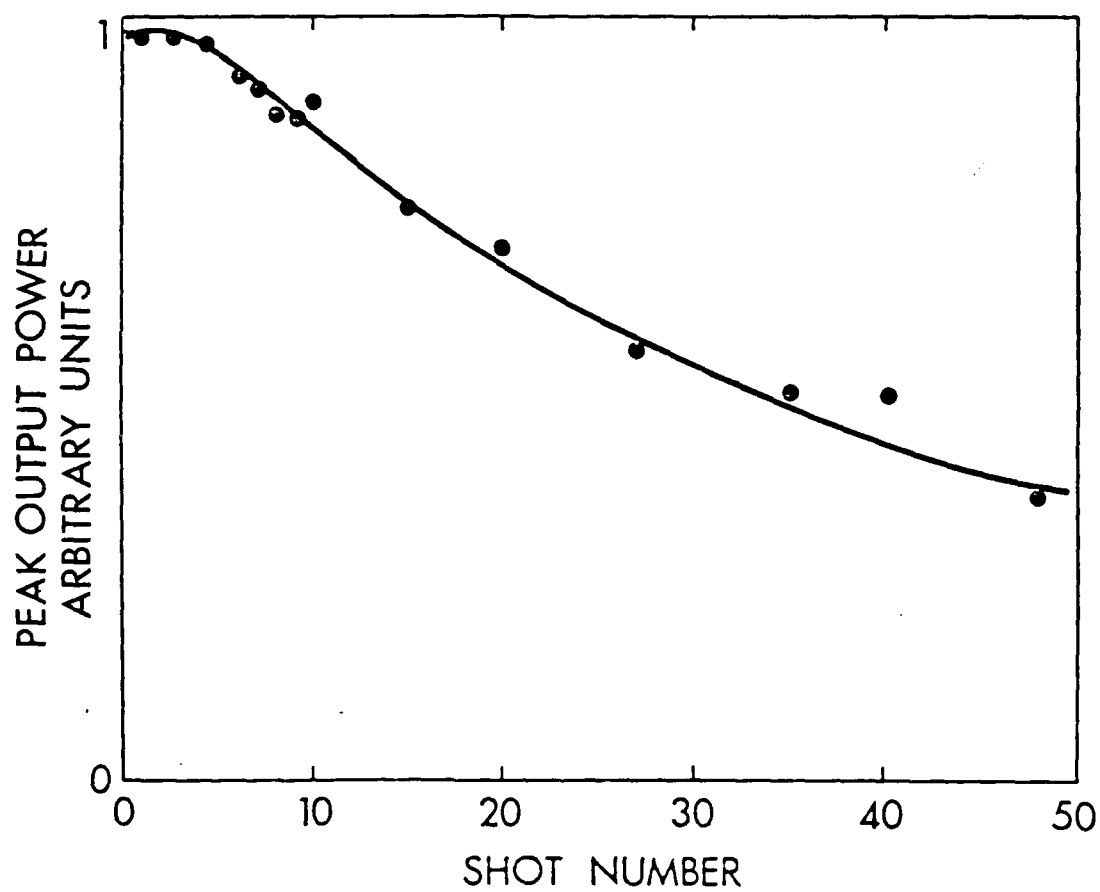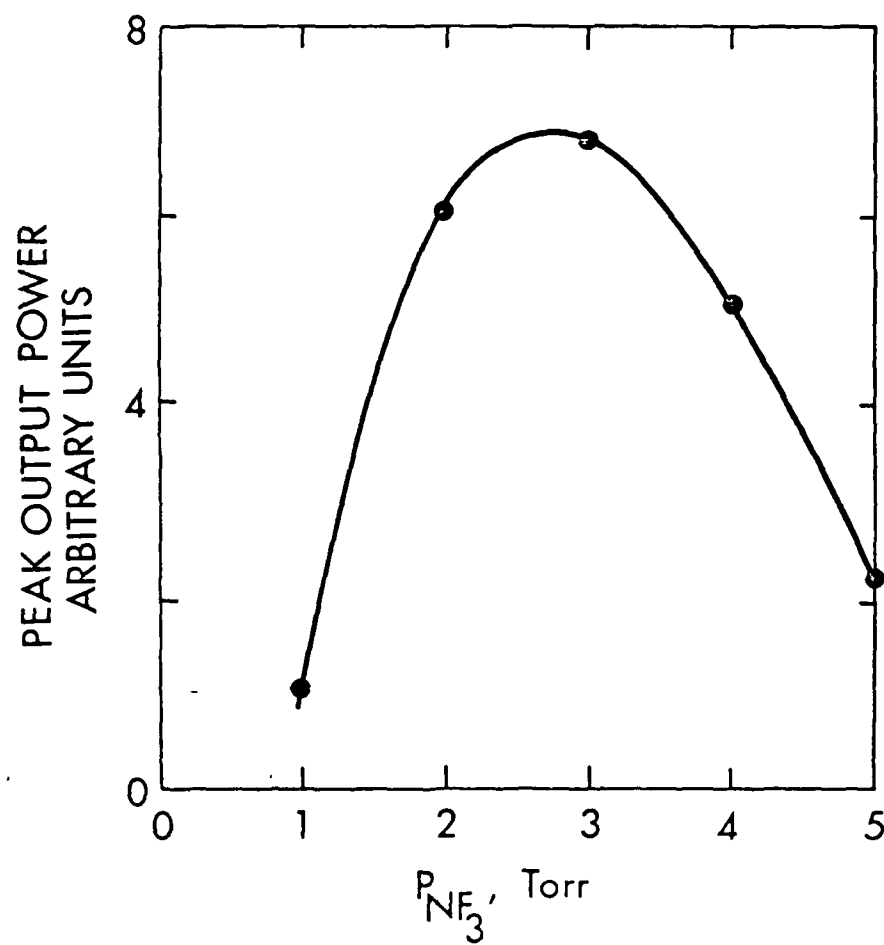
482 486 490 494

470 480 490 500

WAVELENGTH , nm

Fig. 1
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Fig. 2
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Fig. 3.
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PEAK POWER, kW
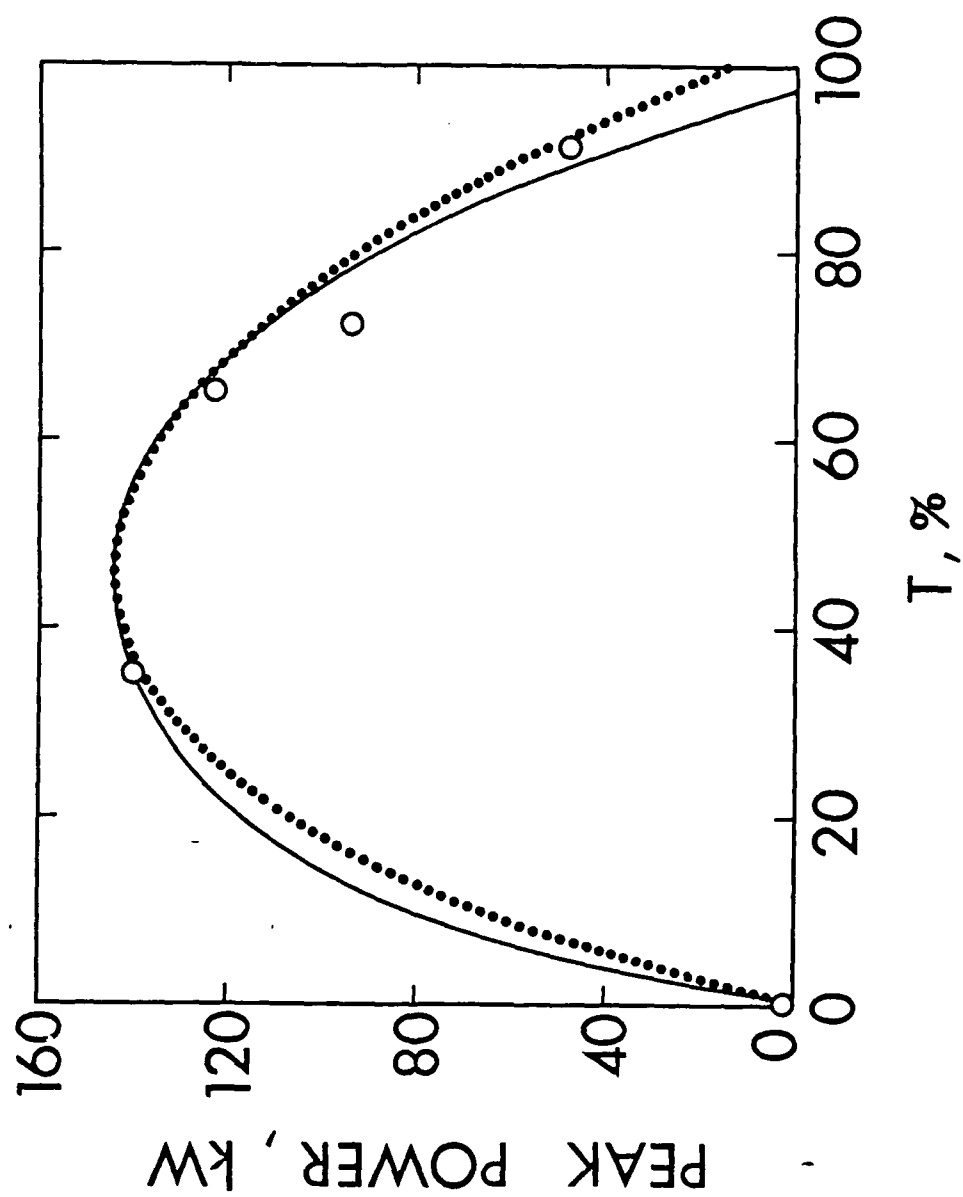
T, %

Fig. 4
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